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REPORT

CD NO.

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COUNTRY	Hungary	DATE OF INFORMATION	1951
SUBJECT	Economic - Chemical industry, natural gas, cracking gas		
HOW PUBLISHED	Monthly periodical	DATE DIST.	4 Jun 1952
WHERE PUBLISHED	Budapest	NO. OF PAGES	21
DATE PUBLISHED	Aug, Sep, 1951		
LANGUAGE	Hungarian	SUPPLEMENT TO REPORT NO.	

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FEB 7 1955

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SOURCE Magyar Kemikusok Lapja, Vol VI, No 8, 9, 1951.

PROBLEMS IN THE EXPLOITATION OF HUNGARIAN NATURAL GAS AND CRACKING GAS

Edited by Dr Gyula Nyul

Natural gas is one of Hungary's most valuable raw materials which, however, is still far from being exploited adequately. Since the country does not possess unlimited quantities of natural gas, it is in the interest of the national economy that the most scientific methods be used in the exploitation and processing of the natural-gas resources. Cracking gas, a valuable by-product of petroleum which finds numerous applications in the chemical industry, can also be derived from natural gas, although in smaller quantities than from petroleum.

The research staff of MAFKI (Magyar Asvanyolaj-es Foldgaz Kiserleti Intezet, Hungarian Research Institute for Petroleum and Natural Gas) is responsible for most experiments connected with natural-gas production. The results of these experiments and their possible application to industry were discussed by researchers and experts at a meeting held under the auspices of the Society of Hungarian Chemists. The meeting was attended by the country's leading experts and by government representatives, including members of the National Planning Office. /Date and place of meeting not given./ The speakers presented an over-all picture of the most important problems concerning the production of natural gas.

In his opening speech, Dr Mihaly Freund, the presiding officer, director of the Hungarian Research Institute for Petroleum and Natural Gas, said among other things:

The use of natural gas in the chemical industry has so far been realized only by those nations which lead the world in technological development: the USSR, England, Germany, and the US. The development of this branch of chemical industry has, until very recently, been neglected in Hungary, Rumania, and Poland, although the raw material is available in these countries.

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Since the expansion of the chemical industry is one of the foremost goals of the Five-Year Plan, the establishment of a natural-gas chemical industry in Hungary is of the greatest importance. Numerous questions have to be considered in connection with this problem, such as economical transportation methods, the separation of pure hydrocarbons from the gas mixture, the production of acetylene, synthesis gas, and hydrogen from natural gas, and the production of organic substances from the gases.

The papers and comments at the meeting are summarized as follows.

Imre Vadas (National Planning Office. In his absence, Tibor Hegedus read the paper)

Recognizing the importance of the chemical industry in Hungary's economy, the revisors of the Five-Year Plan emphasized, aside from the goals set in the original plan, the production of basic materials and the development of the industries producing those materials. The intensified production of basic materials and increased utilization of domestic raw materials will necessitate wider application of modern technology.

It is well known that great advances have been made during the past few years in the discovery of new raw-material occurrences. In the chemical industry, natural gas is one of the most important available raw materials. At present, the location of appreciable quantities of natural gas has been established, and the long-range program of geologic surveying planned for Hungary is expected to reveal further reserves of natural gas. Consequently, it is necessary that effective steps be taken concerning the earliest possible utilization of natural-gas resources. This question is closely connected with another fundamental problem, namely, the reorganization of the Hungarian chemical industry into a uniform industry which can satisfy the needs of every important sector of the national economy.

The processing of natural gas is guided by the composition of the gas in question. The present meeting is concerned especially with natural gas rich in methane content, but very soon, the problems concerning natural gas rich in carbon dioxide content will have to be considered.

The processing of natural gas encompasses many branches of the chemical industry. First of all, acetylene may be produced from natural gas rich in methane, and during the process, considerable quantities of synthesis gas, another fundamental raw material of modern chemical industry, are created. On the other hand, synthesis gas can also be produced in large quantities directly from natural gas. The important basic substances which can be derived from natural gas by modern technological methods are of especial interest, since they will provide the means with which to alliviate the deficiencies still present in the Hungarian chemical industry. Many organic basic materials, such as alcohols, acids, and esters, as well as various solvents, including benzene, carbon tetrachloride, and dichloromethane, as well as basic materials for synthetic industry (formaldehyde, acetone, cyanide, etc.), can be produced from natural gas. The synthesis gas which can be processed from natural gas greatly increases the producing capacity of the synthetic organic industry. It can be used as a base for the manufacture of nitrogen, artificial fertilizers, synthetic motor fuels (Bergius and Fischer-Tropsch method), methanol, etc.

In countries lacking cheap water power, natural gas may also be used for the manufacture of substitute raw materials for the production of acetylene previously produced from carbide. Natural gas also furnished a suitable raw material for synthetic rubber.

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These few examples should suffice to show the wide possibilities of the use of natural gas in the chemical industry. Modern technology has developed numerous methods of processing natural gas. The problem of finding the processes best suited for Hungarian industry can be greatly facilitated by making extensive use of the successful results of experiments carried out in the USSR.

Progress in natural-gas chemistry is important for reducing the country's dependence on imports, especially of nonferrous metals, from the West. The application of up-to-date technological methods will assure the rapid and correct solution of the problems at hand. These methods will also aid in achieving one of the fundamental premises of socialist economic thought. econ- omy.

Transportation of Natural Gas, by Zoltan Cyulay (University of Sopron)

In designing a natural-gas pipeline, three factors enter into the problem amount of gas to be transported, distance, and pressure at point of origin. In case the pressure at the point of origin is known, the problem is simplified and the pipe diameter may be arrived at directly. On the other hand, if the gas first must be compressed at the point of origin, the problem resolves itself into selecting the most economical diameter. Economical operation also depends on the gas pressure during transmission: the lower the pressure, the larger the depreciation per unit of gas transported.

The so-called Transylvanian formula was developed in 1914 by Jeno Guman on the basis of experiments he conducted on the Sarmas-Torda natural-gas pipeline. Starting with the universal differential equation of the flow of gas, Guman deduced a rational formula, in which the factor of pipe friction is a function of the Reynolds number. To facilitate the practical application of the formula, Guman prepared a graph which can be applied to a wide range of varying technical conditions. The calculation of the pipe diameter by application of the rational formula is time consuming, since the friction factor is a function of the desired diameter.

The so-called load factor represents a percentage of total capacity of the gas pipeline. Load factor in the ratio of actual average gas consumption per hour to the maximum hourly capacity of the pipeline. At a continuously operating industrial plant, the load factor may reach 85-90 percent.

It is important that the pipeline should be as short as possible. In the Transdanubian region of Hungary it is possible to lay a pipeline practically in a straight line. Crossing of highways, railroad lines, rivers, and lakes is no obstacle, and can be accomplished through either an overpass or an underpass. For example, the well-known 850-kilometer Saratov-Moscow pipeline, which was built in 2 years and has a capacity of 500 million cubic meters per year, crosses 40 highways and 80 rivers and lakes.

Next let us discuss the transportation of natural gas in Hungary. Petroleum is transported from Zala County to Budapest through an oil pipeline which is too large for current requirements. The weekly production of the oil field does not utilize the full capacity of the line and leaves 50-55 hours per week available for the transportation of natural gas. The gas is fed into the line in part at well pressure, but for the most part through the use of compressors. The pipeline is switched from petroleum to natural gas as follows: a separating plug is placed in the pipe in front of the gas. With the help of this plug the gas -- under high pressure -- forces the remaining petroleum out of the pipe. The maximum starting pressure for the natural gas is 60 atmospheres.

The quantities of natural gas transported from the Zala County oil fields are small; furthermore, the period of productivity of the oil field is shortened when the natural gas is drawn off. Therefore, other gas sources are needed to

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supply the chemical industry. At Ederics (Zala County) water-containing natural-gas reserves were found recently. The exploiting methods best suited for this deposit have not yet been completely developed. However, until other reserves are found, this deposit will have to serve as the basic supply of the chemical industry. It will be easy to transport the gas from the Ederics field economically. Taking into consideration the daily gas requirements and the location of the consumer, an underground pipeline will be designed to carry the gas at well pressure to the chemical plant.

Comments on Zoltan Gyulay's Lecture

1. Comments by Jeno Guman (Transdanubian Petroleum Enterprise)

I have developed two formulas for the transportation of natural gas. The so-called Transylvanian formula, developed on a purely empirical basis has been, and still is being used with great success for calculating the dimensions of natural-gas pipelines in Transylvania. The other formula has been referred to as the "rational formula," because it is based on the most recent achievements of the theory of liquid flow.

In the early 1930s, Nikuradze of the Georgian SSR conducted extensive experiments regarding the inner mechanics of turbulent flow in pipes with a smooth or rough interior surface. Reynolds' experiments and theory have established the basic difference between laminant and turbulent flow, as well as their quantitative interrelationship. Nikuradze also found limiting values for the turbulent flow, which distinguish between the various kinds of turbulent flow. . . indicated by the Reynolds number, the turbulent flow in a rough-surface pipe has two stages. After a transition from laminar flow, the flow continues for a while as if the pipe was smooth. With the increase of the Reynolds number, the flow enters, depending on the interior roughness of the pipe, a transitional stage in which the resistance factor of the pipe is the function of both the Reynolds number and the degree of roughness of the pipe. The influence of the Reynolds number decreases in proportion to the increase in speed. However, depending on the roughness of the pipe, the flow enters, at a limiting Reynold number, the stage at which a permanent resistance factor becomes effective, i. e., a stage in which the flow is independent of the Reynolds number and is only the function of the degree [and type] of the interior roughness of the pipe.

Establishment of this law of the turbulent flow marked a turning point in the interpretation of the mechanics of flow and is, in my opinion, of vast importance for the future.

The "rational formula" of the resistance coefficient, which the Gyulay mentioned, refers to the stage of turbulent flow in which a constant resistance coefficient is exhibited or, as Nikuradze called it, the stage in which the law of the square of velocity is valid. Technically, this law applies to nearly all types of flows. However, the nomogram mentioned by Gyulay also covers the region corresponding to the resistance coefficient of pipes with a smooth inferior surface.

Regarding the load factor, I should like to mention that during my stay in Transylvania, I collected, over a period of many years, data on the load factor of factories, towns, and natural-gas pipelines. However, in this case, the definition of load factor differs from that used by Gyulay since, instead of taking the maximum hourly capacity, I took the actual maximum hourly consumption. The following table shows the load factor of Transylvanian industrial plants and pipelines

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	Load Factor (percent)
Lampblack factories	90-95
Cement factory	66.3
Inorganic chemical plants	40-49
Rolling mill, nail factory	41
Sugar mill	12
Brick and tile factory	53-57
Household consumption in towns	33-35
Pipeline with sugar mill	41.25
Overloaded pipeline	66-71

2. Comments by Jozsef Gloetzer (Enterprise for Designing Chemical Works)

According to Gyulay, economy of operations depends mainly on the quantity of gas transported. Under domestic conditions, gas must be transported in relatively small amounts over fairly great distances. For this reason, a high rate of investment depreciation per kilometer of pipeline may be expected. As a result, transportation cost alone, without the cost of compression, may be placed at 2.5 fillers per cubic meter.

The quality of the natural gas is an important factor in the cost of its transportation. Delivery costs are affected primarily by the amount of cubic meters delivered; however, specific weight is also a factor to be considered. Therefore, the consumption value (amount of components chemically usable, fuel value, etc.) of one cubic meter of gas will be an important factor in actual transportation costs. The factors deciding the consumption value will assume special importance in calculating actual delivery cost of natural gas rich in carbon dioxide content.

Regarding the possibility of a nationwide pipeline system for transporting natural gas, the feasibility of linking such a system with the planned system for distributing gas to industrial plants and towns deserves serious consideration.

3. Comments by Dr Mihaly Freund (MAFKI)

The usual methods for delivering natural gas are the following: (1) through a pipeline; (2) in steel containers at 150-200 atmospheres' pressure; and (3) in special cases, in railroad tank cars at 10-20 atmospheres' pressure. However, there exists a special method, already tried in practice. The natural gas, or rather the methane, is shipped in heat-conserving [insulated] containers, in a liquid state and under atmospheric pressure.

The theory for this method originated in Hungary about 1934, and foreign factories cooperated in finding a practical solution. Before World War II, the method was tried out on a larger scale in Italy.

The transportation and use of natural gas in a liquid state raises several questions: (1) how much more economical is it than transportation in steel containers? (2) weight of filled containers per cubic meter of gas; and (3) loss of gas incurred when it is shipped by this method.

In connection with these questions, the following table presents the more important facts relating to gases shipped in containers.

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Various Containers Used for Storage and Shipping of Gases

	Gas Stored			Container				
Type of Container	Product	Kg/cu M	Max Heating (cal/cu m) Value	Volume (liters)	Wt of Container	Max Gas Content (cu m)	Wt of Filled Cont. (kg/cu m of gas capacity)	Excess Pressure (atm) in Storage
Special steel container of standard dimensions	methane	0.72	9.500	50	50	12	4.17	200
Shipping container of optimum dimension (product of Dalmine, Milan bus company)	methane	0.72	9.500	1,300	950	250-255	3.73-3.00	150-160
Heat-conserving container for liquid gas	methane	0.72	9.500	1.317	580	928	0.62	0.4
Heat-conserving container of optimum dimension for liquid gas	methane	0.72	9.500	3,003	920	2,146	0.43	0.4
Large container for shipping propane-butane (Gas Trading Enterprise)	propane-butane mixture	2.35	30.000	79	37	13	2.76	6
"Dissous" gas container with acetone	acetylene	1.17	14.000	40	70-80	1-5	14-20	15
100 kg calcium carbide, packed in tin drum	acetylene	1.17	14.000	50	105	30	3.5	--

The large containers used by the Gas Trading Enterprise to ship propane-butane have a volume of 79 liters. The weight of the container is 2.76 kilograms per cubic meter of gas, with a capacity of 13 cubic meters of gas at 6 atmospheres' storage pressure.

For "dissous" gas, circumstances are much less favorable. When "dissous" gas is in 40-liter containers in combination with acetone, then at 15 atmospheres' pressure a maximum of 4-5 cubic meters of acetylene can be delivered. The weight of the filled container is 14-20 kilograms per cubic meter of gas. If solid calcium carbide packed in tin drums is regarded as a gas receptacle, even then the weight of the filled container is 3.5 kilograms per cubic meter of gas.

In the heat-conserving 3,000-liter container of optimum dimension 2,146 cubic meters of liquid gas under atmospheric pressure can be shipped when the weight of the filled container is only 0.43 kilogram per cubic meter of gas -- only one tenth of the weight necessary under present shipping methods.

Naturally these figures cannot be compared with the much cheaper transportation via pipeline. Since the building of a pipeline is very expensive, this method of shipping gas is not applicable when many small and widely dispersed consumers have to be supplied. Therefore, the comparison can be made only between shipping in steel containers and the method of shipping methane in liquid form.

Exact figures are available regarding the loss incurred by shipping gas in liquid form. If the gas is in 3,000-liter heat-conserving containers, the hourly loss does not amount to more than 0.28-0.32 percent of the total gas shipped. If transportation is by truck, the escaping gas can be used as motor fuel. This method has actually been used in Italy. However, in truck transportation, 200-liter receptacles incased in vacuum jackets are used, from which the hourly loss is 0.5-0.6 percent of the total gas shipped. These small containers are equipped with a 5-atmospheres safety valve, so that no loss of gas occurs for approximately one hour even if the motor is not running.

Finally, the following may be mentioned regarding the application of this method in Hungary. One truck with 4,000 cubic meters of gas could supply the total daily need, approximately 3,700 cubic meters, of Zalaegerszeg, Szombathely, Koszeg, and Sopron on a 350-kilometer long round trip. Some of these towns have gasworks and a pipeline system, however. A 320-kilometer truck route could supply all the resorts on Lake Balaton, perhaps even including the city of Veszprem. Gyor, which is 200 kilometers from Kerettye (Zala County), could be supplied by one truck and perhaps one trailer. It can be calculated in a similar fashion whether or not this method of transportation could be used to ship natural gas to larger industrial plants.

In trying to arrive at a satisfactory solution to the whole problem, the most interesting detail will be the shipping container itself, since a 3,000-liter Dewar vessel is inconceivable.

4. Comments by <u>Dr Bela Mory</u> (Enterprise for Designing Chemical Works)

Dr Freund mentioned that during the early 1940s experiments with liquid methane were conducted in Italy. Among others, the Italian State Railroads supported these experiments. To obtain one cubic meter of liquid methane, 280-300 liters of methane had to be used. The liquid methane was shipped in metal containers of the Dewar type, with double walls and flask shaped. Each container had a volume of approximately 200 liters and the operating pressure was 5 atmospheres gage. When filled under atmospheric pressure, the container can be closed. For a while, the evaporation of the methane -- due to heating -- increases the pressure, but when maximum pressure is reached, the safety valve of container is activated. As a result of the increase of temperature, the methane disappears from a container of this kind in approximately 500 hours.

As early as November 1940, experimental truck trips were conducted with liquid methane. For a 100-kilometer trip, 10 kilograms of methane were needed, which is the equivalent of 20-21 liters of gasoline. The liquid methane has to be evaporated in a special evaporator. Another method consists of generating the heat needed for evaporation by means of an electrical resistance wire placed in the container.

<u>Various Methods of Processing Acetylene From Natural Gas, by Antal Laszlo (MAFKI)</u>

Researchers have been occupied for a long time with the problem of producing acetylene from hydrocarbons which are cheap and easily accessible. Numerous processes have been found. For a long time, the manufacture of calcium carbide supplied industry with acetylene. However, during the past 10-15 years the chemistry of acetylene was developed, and consequently a new consumer for acetylene appeared: the chemical industry. This fact is mentioned because today the chemical industry uses, or rather could use, considerable quantities of acetylene if it were readily available.

Aside from the so-called classical method of obtaining acetylene from calcium carbid supplied there are three principal methods for producing acetylene: (1) decomposition by heat, (2) by electricity, and (3) oxidation. Variations exist within each method. Two well-known processes for decomposing by heat are the Wulff process and the Ruhrchemie process. A factory in Huels /Germany/ developed on a large scale a method for producing acetylene with the aid of electric arcs. The Schoch process is also used to produce acetylene by electricity. The oxidation method was developed in the ammonia laboratories at Oppau.

Taking into consideration that the calcium carbide method is called the "classic" one and is also the most widely used, the following discussion of technical advantages and economy of the various methods of acetylene production will consist of a comparison of the arc and oxidation methods with the carbide method.

In producing acetylene from calcuim carbide, saturated acetylene is obtained. If the acetylene is used for nonchemical purposes, this process is the most economical. On the other hand, if the acetylene is to be converted into chemical products, the arc and oxidation methods are more favorable. Of these, the oxidation method is preferable under conditions prevailing in Hungary, because it can be employed in plants of any capacity. On the other hand, the electric method is profitable only in large-scale plants.

Aside from the question of economy, several other points have to be considered for purposes of comparison.

1. The acetylene produced by either method has to be concentrated before it can be processed. It does not make an appreciable difference in cost whether the gas to be processed has an 8-9 percent acetylene content (oxidation method) or a 14-15 percent acetylene content (arc method).

2. The advantage of the arc method consists in the fact that the gases can be recirculated and thus the methane is fully utilized. The methane remaining after the oxidation process has to be converted into synthesis gas by additional oxidation.

3. A comparison of the by-products of both methods reveals that the arc method yields numerous by-products, while the by-products of the oxidation method are large quantities of synthesis gas and relatively small amounts of lampblack. As a result, the arc method is applicable only in large-scale operations, where quantities of by-products are obtained and then processed. The separation and processing of the by-products requires difficult operational procedures and an advanced technology. Moreover, separation and processing of the by-products of the oxidation method is simpler. From a chemical viewpoint, the useful gases obtained by the oxidation method are a mixture of acetylene, carbon monoxide, and hydrogen.

4. Considering the amount of power needed, the oxidation method is very definitely more advantageous. It only pays to use the arc method in a large concern, where sufficient quantities of cheap electric power are available. With the oxidation method, only a very small investment is needed for electric power. The economic advantage of using this method is determined by the production and price of oxygen.

It is evident from these few facts that oxidation is the method most suited to the conditions prevailing in Hungary. While the arc method is economical only in large-scale operations, the oxidation method can be used in plants of any size. The small amount of energy needed for oxidation does not have to be electrically generated. The separation and processing of the by-products is better suited to domestic conditions. The synthesis gas which is yielded can be utilized even under present conditions, and can later be put to various uses as domestic technology develops.

When the problem arose in connection with the processing of natural gas, the directorate of MAFKI decided in 1949 that the method best suited to domestic conditions were partial oxidation under conditions of a flame reaction. In 1950, domestic experiments with this method were started under the auspices of MAFKI.

Comments on Antal Laszlo's Lecture

1. Comments by Dr Bela Lanyi (Technical University, Budapest)

Hungary must produce acetylene to aid its synthetic organic industry. In this connection, another method of producing acetylene should be mentioned, namely, decomposing methane on a heated stone checkerboard. Because the equipment required is simple, this method merits study. Considering future synthesis-gas production, the oxidation method shows tremendous advantages over the other processes. The lampblack, a by-product of this process, can be put to good use in Hungarian industry.

2. Comments by Dr Elemer Papp (Institute for Aluminum Research)

Some questions, not answered in the previous lectures, definitely merit further study.

Economy is the decisive consideration in connection with the processing of natural gas. Hungary's natural-gas reserves are limited. In planning the different methods of utilization of natural gas, the basic economic premise should always be that natural gas may be used only for those chemical processes which yield the most needed products.

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Therefore, in discussing the various processing methods, the expenditure of natural gas must be considered, especially with reference to the value of the by-products. The possiblity exists that the requirements of the Five-Year Plans will not emphasize the production of synthesis gas yielded by the oxidation process but will stress the importance of using a large percentage of the available natural gas for the production of purest-quality acetylene, which requires a different process.

In evaluating these tasks from the technological and economic viewpoints, the so-called key value of acetylene obtained from natural gas is important. Key value is a figure indicating the amount of the end products, such as polyvinyl resins, acetone, and acetaldehyde, obtained from the acetylene corresponding to a cubic meter of natural gas.

Another question to be considered is the application of processes to natural gas which will produce materials lacking in Hungary. For example, if nitrogen is present in the gas, a process can be applied which will yield valuable hydrogen cyanide as a by-product.

3. Comments by <u>Bela Galauner</u> (Felsogalla Carbide Plant)

Acetylene production for industries other than the chemical, such as the metal industry, and for lighting purposes is economical only when the carbide process is applied. In his discussion of the various processes, Laszlo did not mention the factors involved in the cost of production. In the carbide process, these factors enter into the cost in the following manner:

	Percent
Cost of current	36
Cost of coke	16
Cost of quicklime	12
Cost of electrode	6
Cost of other materials	9
Wages	7

In a modern Hungarian concern, the processing of one cubic meter of acetylene from carbide-acetylene requires, aside from available domestic raw materials, 10.7 kilowatt-hours, 2.1 kilograms of coke, 3.3 kilograms of quicklime, and 0.15 percent anode material.

In a modern, medium-sized carbide plant with a yearly capacity of 20,000-40,000 tons of carbide production, cost per cubic meter of acetylene will be approximately 6.5-7 forints.

4. Comments by <u>Dr Jozsef Vandor</u> (Research Institute for the Synthetics Industry)

Natural-gas acetylene will represent an advantage over carbide acetylene only if, through increasing the volume of yield, the cost of purification and concentration is reduced. The synthetic industry needs 99.6 percent pure acetylene. The carbon monoxide and hydrogen mixture present in natural-gas acetylene acts as a catalyst poison. Considering the usual life duration of the catalyst, only a maximum 0.4 percent carbon monoxide content is permitted. For this reason, purification is the decisive factor in the production and utilization of natural-gas acetylene. It has been planned to achieve the required degree of purification through high-efficiency sorption. According to

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present figures, this method has only produced 90-percent pure acetylene. Therefore, before it can be used in the synthetic industry, the acetylene must be further purified. This raises the question whether it is correct to use high-efficiency sorption or whether it would not be more advantageous to use a solvent.

The lecturer has presented a one-sided picture regarding the possibilities of processing acetylene. It is because of the cost of purification that it cannot yet be said that natural-gas acetylene is cheaper and better than carbide acetylene.

/Dr Zoltan Szabo (University of Szeged) and Dr Lajos Almasi (Ministry of Mining and Power, Division of Inorganic Chemical Industry) also commented on Laszlo's lecture. Their comments were not included, since they were not of intelligence interest./

Production of Carbon Monoxide-Hydrogen Gas Mixtures From Natural Gas, by Dr Pal Benedek (MAFKI)

/The lecture consists mostly of textbook-type material. No reference is made to domestic conditions./

Comments on Dr Pal Benedek's Lecture

1. Comments by Dr Jozsef Varga (Technical University, Budapest)

The natural gas found at Inke (Somogy County) and east of the Tisza River is rich in carbon dioxide. The possibility exists that natural-gas reserves will be found which contain hydrocarbons diluted with hydrogen. Therefore it is advisable that the study of the synthesis-gas-producing methods should include the study of gases poor in methane and especially of natural gas with a high carbon dioxide content. This is important not only because Hungary's natural gas reserves are limited, but also because the utilization of carbon-dioxide natural gases is an economic and scientific task of Hungary.

For over a year now, catalytic conversion has been studied at the Technical University, Budapest. At a temperature of 850 degrees and with a nickel catalyst, a gas was produced from methane diluted with 66 percent carbon dioxide. It has been established that in the initial gas 14.8 percent of the carbon dioxide remain unchanged, while the remaining gas results in 46.8 percent carbon monoxide and 33.8 percent hydrogen. The calculations show that if the conversion is conducted in the presence of water vapor, it is possible to produce synthesis gas with a 2:1 hydrogen-carbon monoxide ratio. No carbon separation was observed on the catalysts used for conversion. It seems that the same catalysts can be used for several years.

Such studies are of value only if performed with the requirements of a small plant in mind. However, a heat-resisting steel pipe is needed for the building of a small plant. Therefore, this meeting should inquire into the manufacturing possibilities of such a pipe.

2. Comments by Jozsef Gloetzer

The mentioned processes have not yet been tried on an industrial scale in Hungary. Therefore, the necessary data, such as materials needed, etc., have to be taken from published sources.

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In connection with the question of economy, the question of best utilization of natural gas also requires consideration. Should more reserves be discovered, natural gas probably will achieve importance also as a fuel.

One thousand kilocalories of generator gas, the cheapest fuel gas, cost approximately 5 fillers. This would be the equivalent of approximately 40 fillers per cubic meter of 8,000-kilocaloric natural gas.

The mining cost of better-grade lignite is 3.3-3.5 fillers per 1,000 kilocalories. The equivalent of this at 8,000 kilocalories is 26-28 fillers.

The present well price of natural gas is 20 fillers, which seems cheap compared with the prices mentioned above.

[Matyas Keresztes (Pet Nitrogen Enterprise), Lajos Birtalan (National Planning Office), and Dr Jozsef Berty (MAFKI) also commented on Dr Benedek's speech. Their comments were not included, since they were not of intelligence interest.]

Questions Regarding the Separation of Pure Hydrocarbons From Gas Mixtures, by Dr Mihaly Freund

One of the most important factors in the consideration of gases for chemical processing is the composition of the raw material, in this case either natural gas or cracking gas. The following table shows the composition of the natural gas extracted in southern Zala County.

	Percent
Methane	80.2-85.5
Ethane	7.5-9.6
Propane	3.1-5.6
Butane	2.0-2.8
Pentane and heavier	1.8-2.9

The composition of cracking gas, which contains olefins in addition to many other components, presents a more complicated problem. The table below shows the composition of the waste gases gained by the various processes using decomposition by heat. The composition of the gas processed at Pet is also included.

Composition in Volume Percent of Waste Gases
Resulting From Decomposition by Heat

Decomposition by Heat	Pyrolysis	Vapor Phase	Catalysis	At Pet
H_2	3-4	9-14	7-11	3-4
CH_4	12-22	26-40	11-20	20-28
C_2H_6	17-23	6-12	5-26	15-20
C_3H_8	16-27	1-5	10-15	18-20
C_4H_{10}	6-7	1-2	14-27	7-11

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Decomposition by Heat	Pyrolysis	Vapor Phase	Catalysis	At Pet
C_5H_{12}	0-7	0-4		
C_2H_4	6-7	25-32	4-10	5-7
C_3H_6	11-13	10-17	19-35	10-15
C_4H_8	2-4	4-6	11-12	5-8
Total olefins	19-24	39-55	34-57	20-30

The two types of gas mentioned contain only hydrocarbons and perhaps some hydrogen. The separation of the final gases obtained through partial oxidation or by the processing of synthesis gas is much more involved. Those gases contain carbon monoxide and even small quantities of carbon dioxide, nitrogen, and oxygen.

In general, four methods may be employed to separate the hydrocarbon components from natural or cracked gas: (1) fractional evaporation, (2) absorption, (3) adsorption, and (4) chemical absorption.

In Hungary, one of the most timely questions in the field of gas separation is the extraction of the olefins from cracking gases, since the waste gases are currently used only as fuel.

The final gases created by partial oxidation contain little acetylene, and the extraction of that acetylene presents another problem. Furthermore, the processing of the large quantities of petroleum extracted from the fields in southern Zala County is also important. For a long time this petroleum has been utilized in large-scale production of liquid gas. It seems that the existing facilities can be used for the processing of very pure propane, butane, and even pentane and isopetane. Only the production of ethane may present a problem of the Kerettye processing plant, since the ethane concentrates can, at a later time, serve as the raw material for the manufacture of ethylene [?].

Since Hungary does not have a well developed natural-gas chemical industry, only pure ethylene, concentrated propylene-butylene, and amylene are needed. If the ethylene can be extracted with at least 99 percent purity, it can be used for the manufacture of synthetic polymers (polythene). As a concentrate the ethylene may be used for the manufacture of the chemicals derived from ethylene oxide. Propylene-butylene and amylene, and also olefins of higher molecular weight which can be obtained from gasoline, are needed for oxosynthesis, with which experiments are now being conducted for the manufacture of alcohols of higher molecular weight.

The installations at Pet are adequate to produce these chemicals from cracking gas. Amylene, together with its gaseous paraffins and butylene-propylene impurities, can be precipitated from the gases obtained from gasoline by oil absorption in the process of stabilization. The major portion of the washed-out gas will be a propylene-propane, butylene-butane fraction containing ethane-ethylene as in impurity. On the other hand, the primary gas gained through decomposition by heat consists mostly of H_2 CH_4, C_2H_4, and C_2H_6, with impurities of higher molecular weight. By the copper-complex method it would be possible to produce from this gas 90-95 percent ethylene containing only a small amount of propylene-butylene impurity. In the future, should the necessity arise, it will be possible to extract pure propane-propylene, butane-butylene, and ethane-ethylene vapors if more fractionation towers are installed.

High-efficiency sorption appears to be the best method for processing pure acetylene from the final gases resulting after the partial oxidation of methane.

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Comments on Dr Mihaly's Lecture

1. Comments by Jeno Purman (Transdanubian Petroleum Enterprise)

It is in the interest of Hungary's economy that the processes for the separation of hydrocarbons and for the manufacture of pure hydrocarbons be thoroughly studied.

Aside from fractional distillation, separation of hydrocarbons by absorption and adsorption has long been used by large Hungarian firms. Absorption and adsorption are used in processing the rich petroleum from the southern Zala County fields, and also to separate the light paraffin hydrocarbons (especially propane, isobutane, normal butane, and the pentanes) having properties similar to gasoline.

From the complex hydrocarbon mixture, the so-called crude gasoline, liquid gas (propane-butane mixture), and stabilized gasoline are produced by fractional distillation. The next advance must be made in the production of pure hydrocarbon mixtures.

In addition to the four enumerated by Dr Freund, the recently developed heat-diffusion method should be mentioned. It is still in the laboratory stage but has possibilities for actual adoption in large industrial concerns.

2. Comments by Tibor Jancso (Pet Petroleum Enterprise)

Separation of hydrocarbon gases at the plant of the Transdanubian Petroleum Enterprise is effected partly by the absorption method and partly by the adsorption method. Both methods are combined with fractional distillation.

Our cracking gas industry started in 1940. At first, cracking gases were used exclusively to fuel pipe furnaces. In 1943, a fractionation column with 42 bubble trays (designed by Szigeth and Takacsi) was erected at our plant for the processing of stabilization cracking gases and of "moist" gases gained at the stabilization of top-fraction gasoline resulting from the distillation of petroleum. In this column, at 17 atmospheres' pressure, the stabilization cracking gases and the "moist" gases were fractionated into a top and a bottom product. The latter consisted predominantly of C_3 and C_4 gases which had liquefied under the great pressure.

Since the time our cracking plant was forced temporarily to cease operation, the column has been used for the production of special gasoline. At the moment, cracking gas is again used as a fuel. However, this can not continue any longer, since we are wasting unsaturated hydrocarbons which are of great value to the chemical industry. It is therefore necessary to separate these hydrocarbons and put them at the disposal of the industry, especially since our cracking-gas production is increasing.

One of the intermediary steps in the separation of the olefin content, primarily of ethylene, from cracking gases is the copper-ethanolamine method, which has been found very effective.

On the basis of a rather sparingly worded CIOS report, we installed during the past year a small experimental plant utilizing this method, which, in short, operates as follows. Expanded to the required pressure, the cracking gas is led into the gasometer. Subsequently, the cracking gas is drawn off into a gas compressor through a condenser, a vessel containing activated carbon suitable for separating C_5 fractions, and a gas meter, and its pressure is brought to 17-18 atmospheres. From the compressor the gas, which has been

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cooled to 20 degrees centigrade, is introduced through a nozzle into the first half of an absorption column filled with Raschig rings, and a copperethanolamine solution is pumped in from above. The solution, moving downward in the column under the above-mentioned pressure, absorbs in counter current the unsaturated compounds and forms complexes with them. The gases freed from the olefins are removed from the upper part of the column through an automatic pressure-release valve. The solution containing the unsaturated compounds and some gaseous paraffins absorbed during the process goes to the bottom of the column. From there it is conducted into an expansion vessel set at 2 atmospheres' pressure where the absorbed gaseous paraffins are liberated. The solution, together with the olefins, is now conducted into a reheater set at 40 degree. centigrade from which it continues into a vessel placed under vacuum, where at 150-350 millimeters of mercury absolute pressure the unsaturated compounds are liberated from the solution. The unsaturated compounds are then conducted through a 7 percent KOH solution, drawn off with a water vacuum pump /water ray pump?/, and stored as finished gas products.

The solution which remained in the vessel under vacuum is drawn off through a condenser and recirculated. The composition of the fluid is as follows:

	Percent
Copper	7.0
Copper oxide	8.7
Monoethanolamine	31.7
Ammonium nitrate	23.8
Water	28.8

The reaction which occurs during the preparation of the solution is exothermic. Therefore the temperature should not be permitted to rise over 70 degree centigrade. According to the report, during this phase of the process explosions occurred twice at the Leuna Works.

To pervent oxidation through contact with air, we prepared and handled the solution in vacuum. For this reason, the solution was kept under nitrogen or cracking gas.

The most important operating factor in the solution of the gas is pressure. We achieved optimum results at 18 atmospheres' pressure. Below that pressure the absorption of olefins increased rapidly; above that pressure the higher components of the cracking gas liquefy, but the absorption of the olefins does not increase. It is very important to keep the pressure constant. A change in pressure exerts an unfavorable influence on the second important operating factor, the washing solution/gas ratio. According to our experiment, the best result was achieved at 1:30 and 1:50 solution/gas ratio.

The third operating factor is the temperature of solution. We reached optimum results at 20-30 degree centigrade temperature.

In summary, with this method we can extract from the unprocessed cracking gas 90-100 percent of its ethylene content, 80 percent of its total olefin content, and 70 percent of its propylene plus butylene content.

Under conditions prevailing at Pet, it is worthwhile to use the light-gas fraction containing hydrogen, methane, and ethane, through conversion with water vapor, for ammonia synthesis.

In the future, the cracking industry should be gea d to production of high-octane gasolines as well as for the purpose of increasing the quantity of cracking gases, since these gases are becoming an indispensable raw material of the chemical industry.

3. Comments by Benedek Nagy Takacsi (Enterprise for Designing Chemical Works)

The enriching of acetylene is not a simple task since it can not be performed by the usual fractionating methods. Researchers have spent much time on the problem, and Dr Freund has designated high-efficiency sorption as the most efficient method.

Continuous adsorption by way of a moving carbon bed is without doubt an excellent solution. However, we know very little about the practical application of this method. In my opinion, the successful application of this method depends first of all on our ability to produce activated carbon having the necessary firmness. Our domestically manufactured activated carbon has little firmness. Consequently, we must count on a high depreciation rate, which can not be disregarded, since the price of activated carbon is relatively high.

The manufacture of the hyperadsorber (hipersorter), the modern apparatus required for this method, will present a serious problem unless we can avail ourselves of the blueprints. We have relatively little experience in drawing up plans for special apparatus, and we do not have enough personnel to design plans for an unknown type of apparatus. The hyperadsorber is very modern and complicated, and it would take lengthy experimentation and repeated rebuilding before it is available for practical application.

It seems to me that the enriching of acetylene can be performed better by absorption with water. This method has been used on an industrial scale in several places. In this method the cooled gas coming from the combustion equipment contains 8-9 percent acetylene. The gas is then compressed to 18 atmospheres' pressure and washed with water, which absorbs the acetylene and part of the carbon dioxide. According to reports, it is possible to obtain from the resulting solution a gas containing 70 percent acetylene and 28 percent carbon dioxide. This gas can be further purified in a [caustic] lye or alkacid bath.

The apparatus required for this method is well known in Hungary and therefore no problems would arise in equipping a plant. In any case, we have to be careful that no acetylene explosions occur when the gas is compressed. To a smaller extent, the danger of explosion is also present at high-efficiency sorption, because in this process the gas must also be compressed.

4. Comments by Endre Vamos (MAFKI)

Research is being carried on at (MAFKI) regarding the separation of petroleum products by thermo-diffusion. The results of this research show that thermo-diffusion, although very efficient from a theoretical standpoint is not yet applicable on a practical basis, especially not for refining petroleum vapors of high molecular weight. Separation of these complicated mixtures is a very slow process requiring a tremendous amount of energy.

5. Comments by Dr Laszlo Graf (Transdanubian Petroleum Enterprise)

An important factor in the problem of domestic gas utilization, especially of the gases originating on the Great Plain, is the separation of carbon dioxide from the hydrocarbon gases (methane). To enrich the methane content of gases containing carbon dioxide or to extract the carbon dioxide completely is one of the most timely questions of natural-gas utilization.

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At first glance this task, viewed from the technical angle, seems to be easy, since a number of absorption methods are known and have been proven successful. For our purposes, the simplest of these methods, washing with water under pressure, seems to be satisfactory. Separating the carbon dioxide from the hydrocarbon gases is a much simpler task than separating the hydrocarbons from each other.

However, it must be kept in mind that the domestic natural gases are available under high (70-100 atmospheres' gage) pressure, and that their carbon dioxide content is very great (60-70 percent of volume). To find, under these circumstances, the best technical solution for eliminating carbon dioxide from the natural gas is not a routine task.

The washing or solution methods mentioned in the foregoing, which are in general use in industry, employ a substantially lower pressure (maximum 30 atmospheres' gage). Increase of pressure in the washing columns is inopportune, due to the disproportionately high cost of investment for high-pressure equipment (regulators, pumps, etc.). Furthermore, above a certain optimum pressure, operating expenditures also increase, despite the fact that, by expansion of the escaping carbon dioxide in the turbines, 60 percent of the energy required for circulating the washing solution may be recovered.

The pressure of our high-pressure gases containing carbon dioxide therefore must be reduced before purification by absorption takes place. The resulting expansion produces a loss which could be reduced by high-pressure turbines. However, specially constructed turbines cannot be obtained on short notice.

High gas pressure for the removal of carbon dioxide may be employed due to the thermodynamic properties of carbon dioxide. Industrial utilization of this method was recommended 10 years ago and has been advocated repeatedly in the meantime. This solution seems simple if the principle of recuperative cooling is applied, namely, the carbon dioxide gas is under pressure while it is cooled by the expanding gas. During the cooling process, one part of the carbon dioxide liquefies and is separated from the rest of the gas in a separator. According to calculations, the heat required for distillation of the liquid carbon dioxide is sufficient for producing the cooling effect needed for liquefaction. Thus cooling and enriching of the hydrocarbon gases can proceed without, external interference.

Aside from theoretical calculations, the experiments conducted last year also indicate that this method may be employed for the purification of carbon-dioxide gases. Thus, for example, under 50 atmospheres' gage pressure at -50 degrees centigrade the gas from the Kor[illegible]szegapati (Bihar County) deposit separates into a gas rich in hydrocarbons and a liquid rich in carbon dioxide. Under the above circumstances (50 atmospheres' gage pressure at -50 degrees centigrade) the Orsat apparatus shows the following approximate mean values for the composition of the samples taken from the top and bottom of the separator, respectively:

	Top Sample (gaseous state)	Bottom Sample (liquid state)
	In Volume Percent	
Carbon dioxide	19.0	83.0
Oxygen	1.0	0.2
Hydrocarbon gas	61.0	13.0
Nitrogen	19.0	3.8
	100.0	100.0

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Tentative experiments indicate that a large part of the hydrocarbon gas is dissolved in the carbon dioxide and is lost. However, the process enables us, by distillation under pressure, to utilize the differences between the boiling points of hydrocarbon gas and carbon dioxide. Experiments show that combustible gas may be produced by this method even from carbon dioxide with 2-3 methane volumes.

However, practical application of the deep cooling technique under pressure is not simple. Particularly difficult problems arise in connection with the elimination of the disturbing effect of water and the establishment of smooth operations in the shop.

6. Comments by Dr Jozsef Vandor

While in principle I do not object to high-efficiency sorption, I do not believe that the date compiled by MAFKI insure a satisfactory practical application of this method.

The speedy realization of the suggestion made by MAFKI, namely, that the synthetic industry utilize the dilute unpurified acetylene directly, is quite impossible from the industry's point of view. This is just an attempt to dump the difficult problem of acetylene enriching and purification in the synthetic industry's lap. As far as we are concerned, the acetylene must continue to be purified before we can use it.

7. Comments by Dr Pal Benedek

According to Benedek Nagy Takacsi, there are substantial difficulties in connection with introducing the high-efficiency sorption method in Hungary, because we have not had any experience with this method. However, I do not share his pessimism. Partial oxidation also is a new method, but nevertheless we shall introduce it. Introduction of this new technology in Hungary is predicated on the following factors. (1) the entire literature and industrial history of the method are known to us; (2) we are in possession of substantial experimental material; and (3) Soviet experience, especially the theoretical and practical advices of Soviet Academician Dubinin, are at our disposal.

Production of Organic Substances From Natural Gas and Cracking Gas, by Dr Jozsef Berty

The so-called petrochemical industry, based on the exploitation of petroleum and natural gas, is developing into a new industry in Hungary. Before entering into details, the following basic questions should be reviewed:

As to the raw materials, we possess a large, but not unlimited quantity of natural gas. Our cracking plant is small and cannot satisfy all of the olefin requirements.

As to methods, the new industry should be built up gradually. The substances should be produced from a single or from a few chemical compounds, thereby assuring economical production. Petrochemistry is faced with the same problems as organic chemistry, namely, that while a reaction may be accomplished in one way or another, the most difficult and costly task is the isolation of the product. Application of the so-called destructive methods is economical only at an annual production of several thousand tons. Moreover, the products are seldom used in the same proportion in which they produced.

As to the products, it is important to manufacture first the materials which are required in the largest quantities, and serve for further processing. These materials are:

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C_1	methanol (formaldehyde)
C_2	acetylene, ethylene
C_3	acetone
C_3-C_{20}	aldehydes and alcohols, perhaps acids
C_4	isobutylene, butadiene

C_1

Formaldehyde may be produced either from methane directly or over synthesis gas and methanol. I believe that we should choose the latter method. We need as much methanol as formaldehyde. A large methanol plant is more economical. We already have a small methanol-formaldehyde plant and are familiar with the technology. Synthesis gas is available as a by-product of acetylene manufacture.

C_2

On the basis of the preceding discussion, it has become clear that C_2 hydrocarbons should be produced from acetylene instead of from ethylene.

C_3

The most important basic material for the C_3 compounds is acetone. The most widely used method, the hydration of cracking-gas propylene into isopropyl alcohol and the dehydrogenation of the latter into acetone, is impossible, since the domestic cracking gas does not yield enough propylene. Acetone may be produced directly from the final gas resulting from the partial oxidation of methane not previously converted to acetylene.

C_3-C_{20}

In our opinion, aldehydes and alcohols should be produced by the so-called oxosynthesis. The quantity of olefins required for this purpose is available in the cracking gas, gasoline, and gas oil originating at Pet, while the necessary amount of pure water gas may be obtained from the CO derived from ammonia synthesis. Among these alcohols C_3 - C_6 will satisfy requirements for solvents; C_7 - C_{10}, for softeners; and C_{11} - C_{20}, for fatty alcohols.

The following chart shows the steps in the processing of cracking gas.

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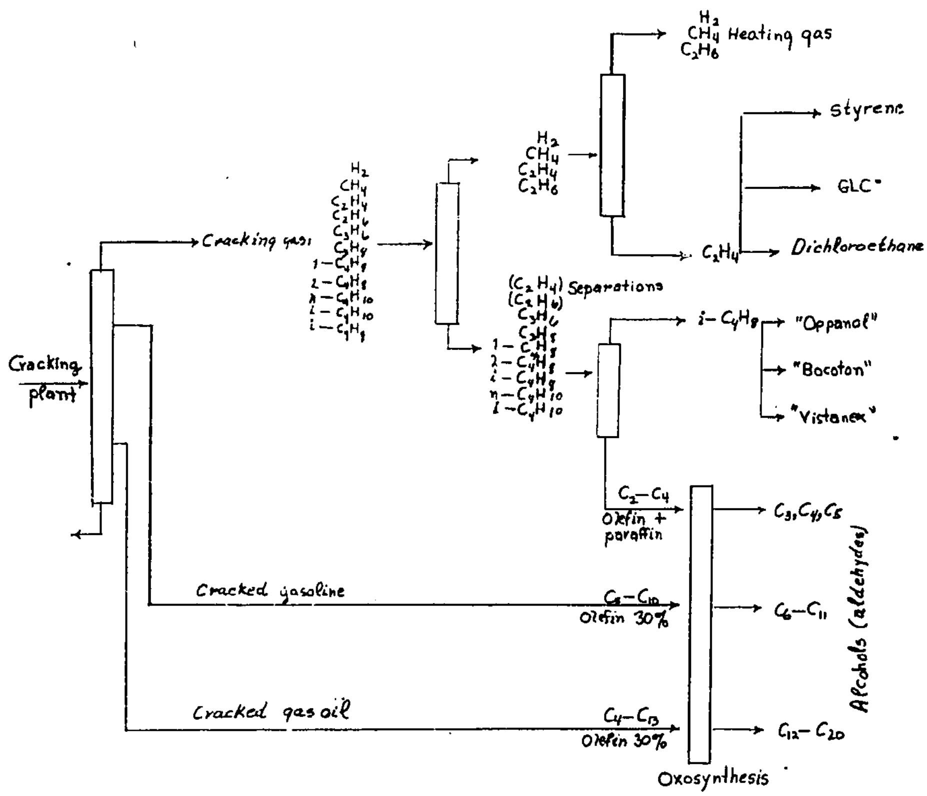
Processing of Cracking Gas



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C_4

[This part of the lecture consists of the presentation of internationally known methods for the production of isobutylene and butadiene without reference to domestic conditions.]

Comments on Dr Jozsef Berty's Speech

1. Comments by Zoltan Szabo

In connection with the manufacture of chlorinated solvents, the question has been raised whether the required amount of chlorine is available. The fact is that, contrary to the situation in countries with a highly developed chemical industry, our own problem is the utilization of the large amount of chlorine orginating as by-product in the manufacture of caustic soda. Caustic soda production represents a bottleneck in Hungarian industry. The Five-Year Plan provides for the creation of additional caustic soda plants. At the same time, an outlet for the resulting increased production of chlorine must be found. One of the possible uses of chlorine is the chlorination of the hydrocarbons of natural gas.

[Also commenting on Dr. Jozsef Berty's speech were: Dr Zoltan Csuros (Technical University, Budapest), Dr Jozsef Vandor, (researcher in synthetics), Dr Laszlo Beregi (Research Institute for the Organic Chemical Industry), Kalman Matolcsy (Research Institute for the Organic Chemical Industry), Dr Endre Zalay (chemical engineer), Dr Janos Schon (Research Institute for the Synthetics Industry), Janos Krepuska (Research Institute for the Synthetics Industry), Ervin Varnagy (Research Institute for the Synthetics Industry), and Tibor Hegedus (National Planning Office). Their comments were not included, since they were not of intelligence interest.]

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